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**Low Loss Second-Order Nonlinear Optical Polymers Based on All Organic  
Sol-Gel Materials**

**by**

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# **Low Loss Second-Order Nonlinear Optical Polymers Based on All Organic Sol-Gel Materials**

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## **SYNOPSIS**

A new class of all organic sol-gel second-order nonlinear optical (NLO) materials based on hexa(methoxymethyl)melamine (HMM) has been developed. Two NLO active chromophores, 4-(4'-nitrophenylazo)aniline (DO3) and 4-amino-4'-nitrobiphenyl (ANB), were incorporated into the melamine matrices. The samples exhibit second-order optical nonlinearity after poling and curing at 220 °C for 30 min. DO3/HMM and ANB/HMM samples show a second harmonic coefficient,  $d_{33}$  of 10.7 and 1.8 pm/V at 1064 nm, respectively. The temporal stability of both systems has been studied at room temperature as well as at 100 °C. Waveguide optical losses of samples at 633 and 830 nm are reported.

**Keywords:** nonlinear optics, organic sol-gel, melamine.

## INTRODUCTION

The development of practical materials for second-order nonlinear optical (NLO) applications, such as frequency doubling and electro-optic (EO) modulation, requires simultaneous optimization of their properties. Large second-order optical nonlinearity, its long-term stability at elevated temperatures, and low optical losses are the most desirable features of materials for such types of applications.<sup>1,2</sup> A number of polymeric materials have been reported to surpass both the second harmonic and linear EO coefficients of the traditional inorganic NLO materials such as lithium niobate and potassium dihydrogen phosphate.<sup>2</sup>

Second-order NLO properties are present in the polymer when the NLO chromophores are aligned in a noncentrosymmetric manner by the poling technique.<sup>3</sup> In order to prevent the randomization of the poled (aligned) NLO molecules in a polymer matrix, NLO chromophores are usually incorporated into a polymer which has a high glass transition temperature ( $T_g$ ). This is due to the fact that the molecular motions of NLO chromophores are closely associated with the  $T_g$  of the polymer.<sup>4</sup> Further, enhanced temporal stability of second-order NLO properties in a poled polymer can be obtained when a certain degree of crosslinking is introduced.<sup>5-14</sup> Long term stability of second-order nonlinearities in many different crosslinked polymeric systems at temperatures higher than 100 °C has been reported earlier.<sup>6,10-13</sup>

Polymeric materials to be used in EO modulators should possess high EO coefficients and low optical losses at their operating wavelengths<sup>2</sup> (typically from 0.8 to 1.5  $\mu\text{m}$ ). In the case of frequency doubling devices, design considerations should seek to minimize the absorption loss at the doubled frequency. Even small absorptions can cause significant damage to the materials.<sup>15</sup> Therefore, a low

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optical loss polymer is an essential requirement in both frequency doubling and EO modulation applications.

Melamine-based materials have been widely used in the lighting, coating, and decorating industries because of their good transparency.<sup>16</sup> The formation of melamine materials via a sol-gel process involves sequential hydrolysis and condensation reactions. This sol-gel process provides an attractive route to the preparation of a densely crosslinked network. In addition, the high  $T_g$  of crosslinked melamine-based materials<sup>17</sup> would be expected to stabilize the aligned NLO chromophores by decreasing their mobility. Therefore, melamine-based polymers with low optical loss, high  $T_g$  and crosslinked features are good candidates for second-order NLO materials.

In this paper, we report the first complete study on an all organic sol-gel second-order NLO polymer. The synthesis and characterization of a melamine-based NLO material is described. Two NLO dyes with different absorption characteristics were added to melamine-based prepolymers. After thermal curing and poling, optically clear crosslinked melamine films containing NLO moieties were obtained. The linear and nonlinear optical properties of the poled/cured samples were determined.

## EXPERIMENTAL

Hexa(methoxymethyl)melamine (HMM, Resimene<sup>®</sup> 747, Figure 1) was obtained from Monsanto and was used as received. HMM is a viscous liquid by nature and is soluble in most common organic solvents. Prepolymer of HMM ( $T_g = 2$  °C) was prepared by heating the monomer at 220 °C for 1 h in the presence of an acid catalyst. Disperse Orange 3, 4-(4'-nitrophenylazo)aniline (DO3, Figure 2a),

is available from Aldrich and was used as received. The other NLO dye, 4-amino-4'-nitrobiphenyl (ANB, Figure 2b), was synthesized according to the literature procedure.<sup>18</sup> The chemical structure was confirmed by IR and <sup>1</sup>H NMR spectra.<sup>19</sup>

Melamine-based samples doped with either DO3 or ANB were further investigated and are hereafter referred to as HMM/DO3 and HMM/ANB, respectively. To prepare a prepolymer solution for spin-coating, an NLO dye and HMM prepolymer in a weight ratio of 1:5 were dissolved in propylene glycol methyl ether acetate (PGMEA) with a certain amount of water and acid catalyst. In one example, DO3 (0.1g) and HMM prepolymer (0.5 g) were dissolved in 4.0 g of PGMEA. The solution also contained 0.6 g of water and 0.3 g of acetic acid to aid the hydrolysis of the HMM prepolymer. This solution was stirred for 1 h at room temperature and then refluxed for 1 h before spin-coating. Films were prepared by spin-coating the solutions onto appropriate substrates. The curing condition for both materials was chosen to be 220 °C for 30 min, partly due to the fact that the thermal degradation temperatures ( $T_d$ ), determined by a thermogravimetric analyzer (TGA), of DO3 and ANB were 250 and 253 °C, respectively. Typical thicknesses of the cured HMM/DO3 and HMM/ANB samples obtained were approximately 0.48 and 0.60  $\mu\text{m}$ , respectively. Indices of refraction at two different wavelengths (532 nm and 1064 nm) were measured using an ellipsometer (Rudolf Research). The indices of refraction for the cured HMM/DO3 sample are 1.709 and 1.668 at 532 and 1064 nm, respectively. For the cured HMM/ANB sample, they are 1.569 and 1.551 at 532 and 1064 nm, respectively.

The  $T_g$  and the reaction behavior of the materials were studied using differential scanning calorimetry (DSC, TA Instrument DSC2910) at a heating rate of 10 °C/min. The thermal degradation temperatures ( $T_d$ ) of the polymers

were determined on a TGA (TA Instrument TGA2950) with a heating rate of 10 °C/min under air. UV/Vis spectra were recorded on a Perkin Elmer Lambda 9 spectrophotometer. Measurements of the dielectric properties of the polymers were performed with a Hewlett Packard model 4284A multifrequency LCR meter. The details of the experimental set-up have been described earlier<sup>20</sup> Samples were measured from 30 °C to 150 °C on a computer controlled hot stage with a constant heating rate of 2°C/min. Optical waveguide losses were measured at 633 and 830 nm. The experimental set-up is described in the literature.<sup>21</sup>

The corona poling technique was used to impart second-order NLO properties in the polymer films. The details of the corona poling set-up have been reported earlier.<sup>6-8</sup> Poling process was initiated at 60 °C and the temperature increased up to 220 °C at a rate of 10 °C/min. The corona current was maintained at 3  $\mu$ A while the poling temperature was kept at 220 °C for 30 min. The formation of the crosslinked network and the alignment of chromophores was carried out simultaneously during this period. The sample was then cooled down to room temperature under the influence of the poling field.

The second-order NLO properties of the poled HMM/DO3 and HMM/ANB samples were measured by second harmonic generation (SHG) from 1064-nm laser radiation. The relaxation behavior of the second-order NLO properties was studied by monitoring the decay of the effective second-order NLO coefficient,  $d$ , as a function of time at both room temperature (25 °C) and 100 °C. The measurements of the second harmonic coefficient,  $d_{33}$ , have been previously discussed<sup>6,13,22</sup> and the  $d_{33}$  values were corrected for absorption.<sup>7</sup>

## RESULTS AND DISCUSSION

The  $T_d$  of the cured HMM/DO3 and HMM/ANB, as taken from the onset point of the step transition, were 278 and 280 °C, respectively. The thermal behavior of the HMM/DO3 sample was studied by DSC. An exothermic reaction (curing) starting at approximately 200 °C was observed. A higher curing temperature is necessary for high  $T_g$  systems due to a slow reaction rate after vitrification. However, curing and thermal degradation often compete at high temperatures.<sup>23</sup> Further, in order to obtain second-order NLO polymer thin films, curing and poling are conducted simultaneously. When the poling temperature is high, ionic conductivity adversely affects the alignment of the NLO chromophores.<sup>24</sup> These factors were taken into consideration in optimizing the curing conditions. Based on the above criteria, isothermal heating of the sample at 220 °C for 30 min was chosen as the optimum curing conditions.

On heating the HMM/DO3 sample at 220 °C for 30 min, a broad transition temperature near 100 °C was observed from the DSC thermogram (Figure 3). A similar  $T_g$  was observed for the HMM/ANB sample. A thin film of cured HMM/DO3 sample was soaked in PGMEA (a good solvent for both HMM and DO3) for 8 h. The solvent did not extract any measurable amount of dye from the cured sample as determined by UV/Vis spectroscopy. This confirmed that a highly crosslinked polymer network incorporating NLO chromophores was formed after the curing process. The cured materials exhibited dielectric constants ( $\epsilon'$ ) in the range of 3 to 4 at room temperature in the frequency range from 30 to 1 MHz. To further investigate the relaxation behavior of these polymers, the dielectric dissipation factors ( $\tan \delta$ ) of pristine and cured samples were compared. Figures 4 shows the  $\tan \delta$  values for the pristine and the cured HMM/DO3 samples as a function of temperature at three different frequencies.



For pristine HMM/DO3, the onset temperature of the  $\tan \delta$  values appeared in the vicinity of 60 °C. After curing the sample at 220 °C for 30 min, the  $\tan \delta$  curves shifted approximately 20 °C higher. The  $\tan \delta$  values increase as the temperature increases and did not show any relaxation peak up to 150 °C, the highest temperature for which measurements were performed. The mobility of the NLO chromophore is expected to be restricted at temperatures lower than 100 °C, as implied by the small  $\tan \delta$  values observed for cured HMM/DO3 sample. As mentioned earlier, the  $T_g$  of the optimally cured HMM/DO3 is 100°C. These results further confirm that the onset of relaxation curves from the dielectric spectra is directly related to the  $T_g$  of this polymer.

The poled/cured films of HMM/DO3 and HMM/ANB exhibit  $d_{33}$  values of 10.7 and 1.8 pm/V respectively at the 1064-nm fundamental. The larger  $d_{33}$  value of the HMM/DO3 than that of the HMM/ANB is because the product of the dipole moment ( $\mu$ ) and the first hyperpolarizability ( $\beta$ ) of DO3 is larger than that of ANB.<sup>25</sup> Figures 5 and 6 show the temporal stability of second-order nonlinearity for poled/cured samples at both 25 and 100 °C. The results clearly indicate that the poled/cured HMM/DO3 showed better stability than HMM/ANB. The effective second-order NLO coefficient  $d$  of the poled/cured HMM/DO3 remained unchanged at 25 °C for as long as the stability was monitored (40 h). In contrast, a reduction of 17% in  $d$  for the poled/cured HMM/ANB was observed under the same condition. Moreover, when the samples were subjected to 100 °C for 40 h, a reduction of 43% in  $d$  was observed for the poled/cured HMM/DO3. In the case of HMM/ANB, a reduction of 55% in  $d$  was observed under the same thermal treatment. The larger molecular size of DO3 compared to that of the ANB accounts for the better temporal stability of this system. It is noted that the second-order optical nonlinearity in both samples showed a fast initial decay followed by a slow decay at 100 °C. Although results from DSC and

dielectric measurements showed an  $\alpha$  transition ( $T_g$  like motion) at around 100 °C, the materials exhibited quite stable nonlinearity at such high temperatures. The enhanced stability at temperatures close to  $T_g$  is attributed to the presence of the crosslinked network in this system.

UV/Vis spectroscopy was used to investigate the absorption behavior of the HMM/DO3 and HMM/ANB samples. Figure 7 shows the absorption spectra for a HMM/DO3 sample before and after poling/curing as well as for the sample that was heated at 100 °C for 40 h. Immediately after poling/curing, a decrease in absorbance was observed as expected. This is due to some dye degradation (or sublimation) and orientational dichroism.<sup>26</sup> The absorption peak of the DO3 chromophore shifted slightly toward shorter wavelength. This behavior is similar to that observed in other crosslinked NLO polymer systems.<sup>7,12</sup> After the sample was heated at 100 °C for 40 h, the absorption spectrum showed only a slight change (see Figure 7). In contrast, the HMM/ANB sample showed an increase in absorbance at the  $\lambda_{\max}$  after thermal treatment (Figure 8). This is a result of the relaxation of the poled order. The observation from Figures 7 and 8 also confirms the decay behavior of the nonlinearities in both samples.

The optical loss of polymer waveguides for HMM/DO3 and HMM/ANB was determined by measuring the scattered light along the guided streak, which is based on the assumption<sup>27</sup>,

$$I = I_0 \exp (-\alpha x) \quad (1)$$

where  $I$  is the intensity scattered from the waveguide at any point,  $I_0$  is the initial intensity at  $x = 0$ , and  $\alpha$  is the attenuation coefficient. An optical fiber probe was employed to measure the light scattered from the waveguide. The scattered light intensity was measured as a function of the probe position. The optical loss data

are listed in Table I. It is important to point out that the condensation of the prepolymer HMM may generate water and alcohol. However, no microscopic void due to water or alcohol generation was observed in the cured thin films of both materials by optical microscopy. Waveguide loss measurements established that the materials suffered reasonably low loss for a crosslinked system. Further optimization of materials and processing conditions is expected to improve the optical loss of the polymers.

## CONCLUSIONS

Organic sol-gel melamine-based polymeric materials for second-order nonlinear optics have been synthesized. The  $T_g$  of these materials was observed at around 100 °C. Two poled/cured melamine-based materials incorporating DO3 and ANB NLO chromophores with second harmonic coefficients,  $d_{33}$ , of 10.7 and 1.8 pm/V were obtained. These samples exhibit a slow decay of the optical nonlinearities over 40 h at 100 °C. The materials have also shown low optical losses at 633 and 830 nm. Optical loss at longer wavelengths, such as 1.3  $\mu\text{m}$  is expected to be even lower. These materials are good candidates for the fabrication of optical waveguide devices.

Partial funding from ONR is gratefully acknowledged.

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19. After recrystallization from a hot, saturated methylene chloride solution, a bright orange powder product was obtained. IR (KBr): 3490  $\text{cm}^{-1}$ , 3385  $\text{cm}^{-1}$  (N-H stretch); 1629  $\text{cm}^{-1}$  (N-H Bend); 1496  $\text{cm}^{-1}$  ( $\nu_{\text{assym}} \text{N=O}$ ); 1332  $\text{cm}^{-1}$  ( $\nu_{\text{sym}}$ ); 826  $\text{cm}^{-1}$  (C-N stretch);  $^1\text{H}$  NMR (DMSO- $d_6$ ):  $\delta$  5.64 (s, 2H), 6.72 (d, 2H), 7.56 (d, 2H), 7.80 (d, 2H), 8.24 (d, 2H); UV-Vis (Methanol): 247 nm, 371 nm ( $\lambda_{\text{max}}$ ), 500 ( $\lambda_{\text{cutoff}}$ ).
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## Captions

**Figure 1** Chemical structure of HMM.

**Figure 2** Chemical structures of (a) DO3 and (b) ANB.

**Figure 3** DSC thermograph of the cured HMM/DO3.

**Figure 4** Temperature dependence of  $\tan \delta$  for pristine and cured HMM/DO3 at different frequencies.

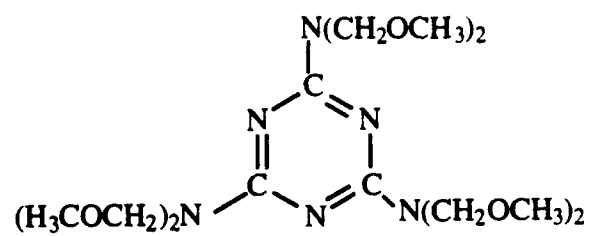
**Figure 5** Temporal behavior of the effective second-order NLO coefficient of the poled/cured HMM/DO3.

**Figure 6** Temporal behavior of the effective second-order NLO coefficient of the poled/cured HMM/ANB.

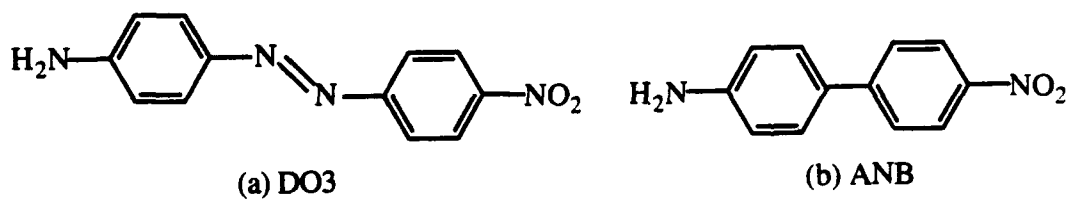
**Figure 7** UV/Vis absorption spectra of the HMM/DO3: (a) pristine, (b) immediately after poling/curing, (c) poled/cured sample, thermal treatment at 100 °C for 40 h.

**Figure 8** UV/Vis absorption spectra of the HMM/ANB: (a) pristine, (b) immediately after poling/curing, (c) poled/cured sample, thermal treatment at 100 °C for 40 h.

**Table I** Optical loss data for the cured HMM, HMM/DO3, and HMM/ANB.

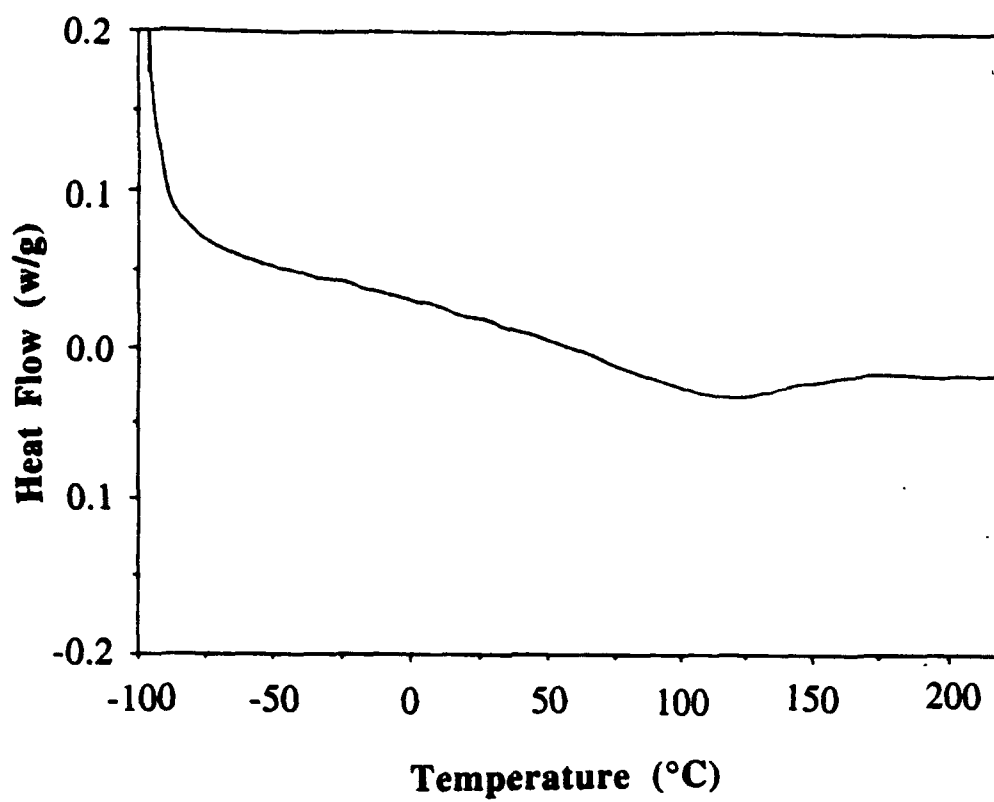


(Figure 1)

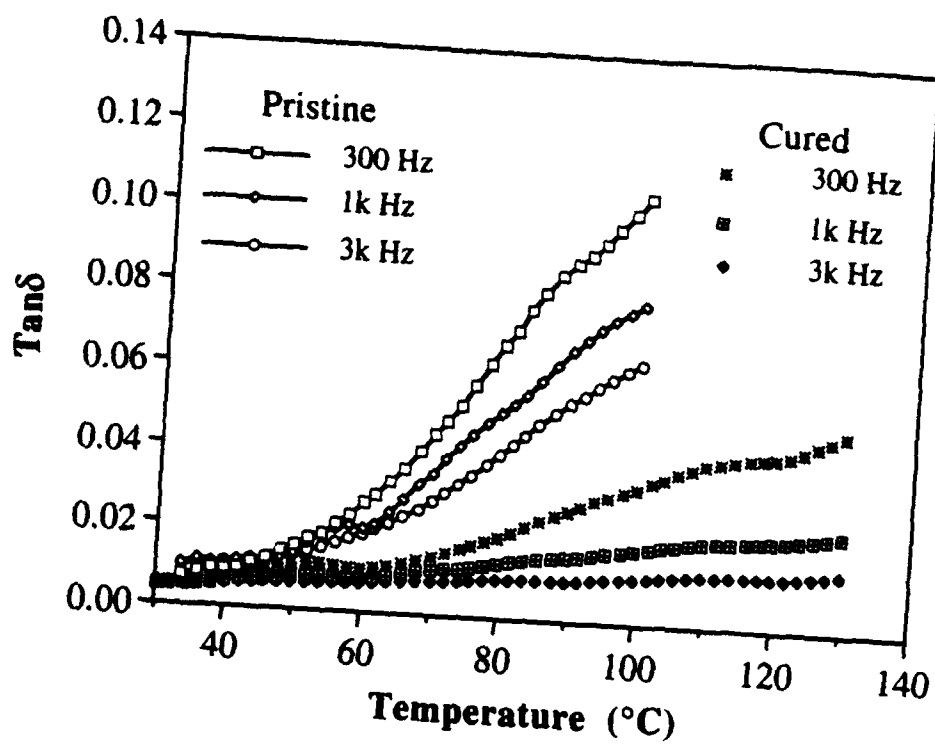


(Figure 2)

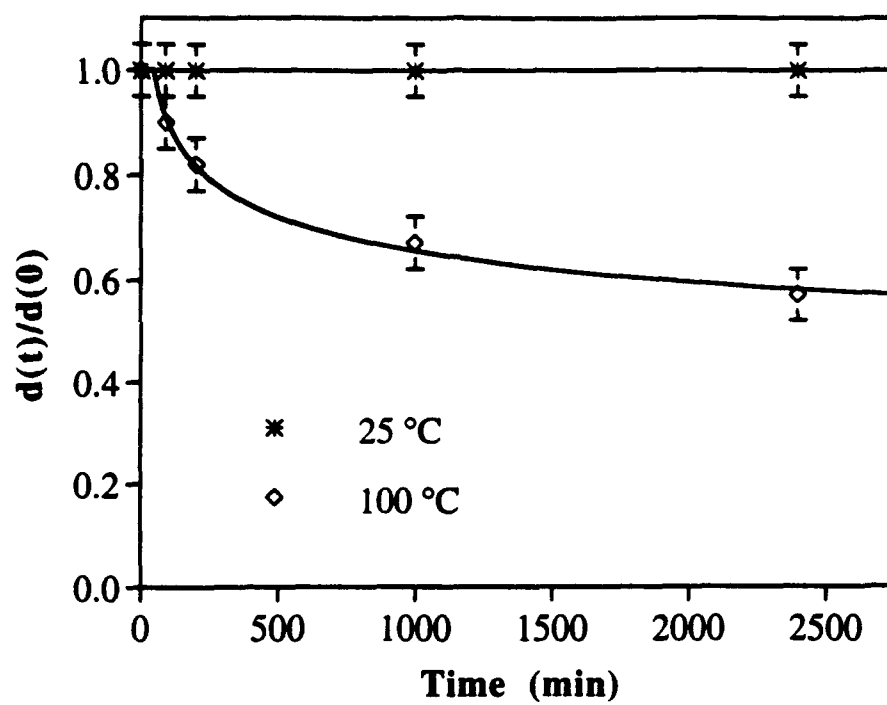




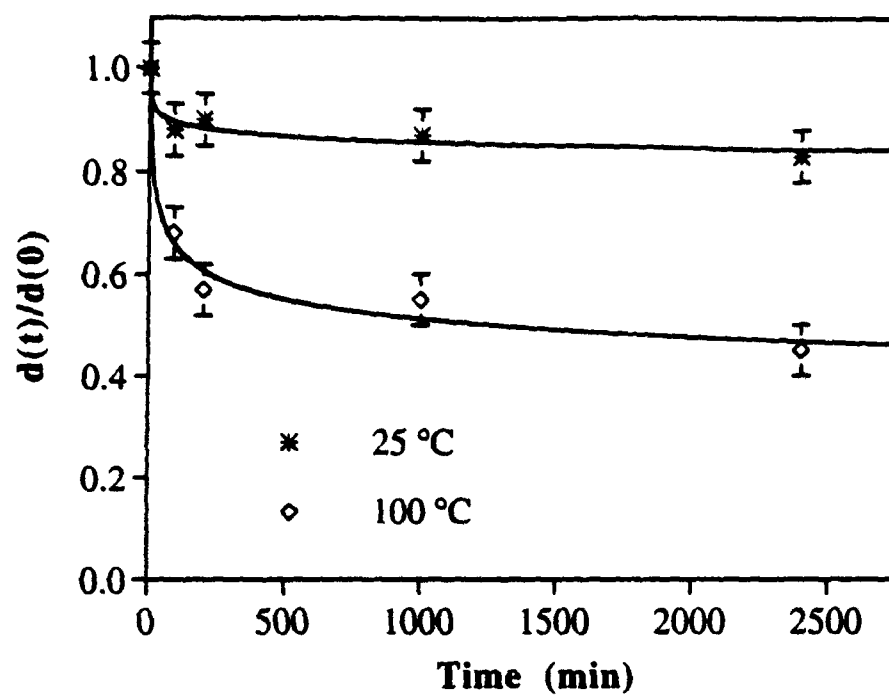
(Figure 3)



(Figure 4)



(Figure 5)



(Figure 6)

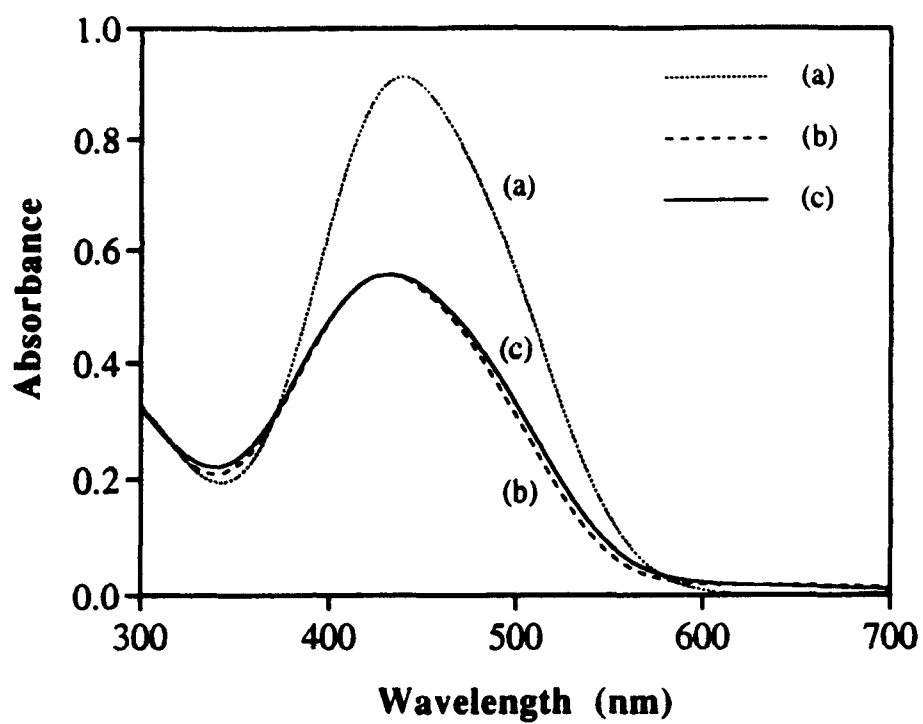
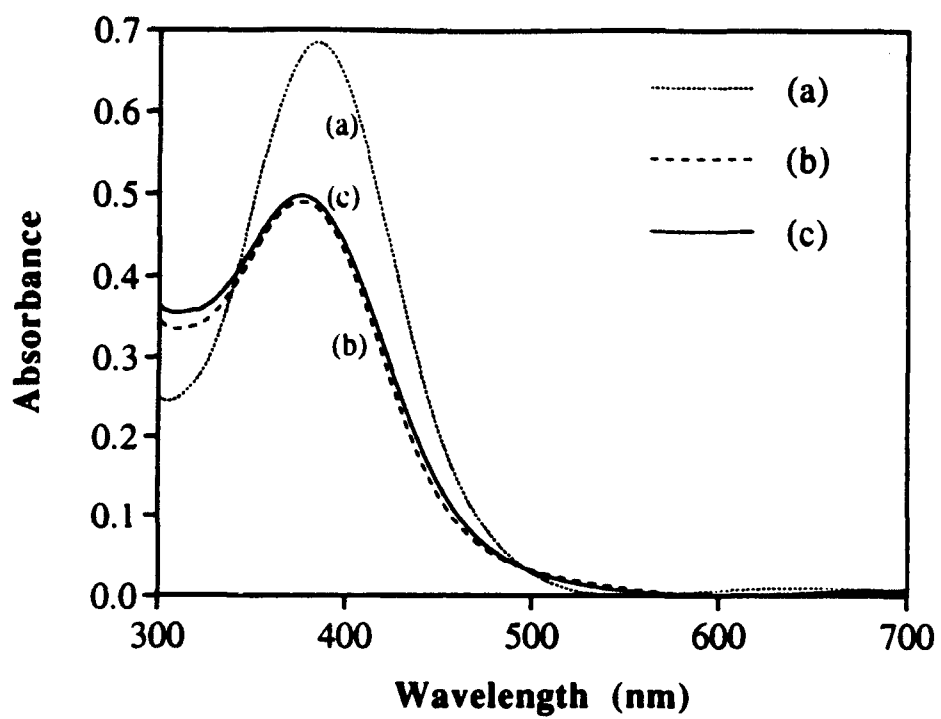


Figure 7.



(Figure 8)

	HMM (dB/cm)	HMM/DO3 (dB/cm)	HMM/ANB (dB/cm)
633 nm	3.0	—*	8.0
830 nm	2.1	3.7	3.3

\*Optical loss was not determined due to the absorption tail of DO3.

(Table I)

**END  
FILMED**

DATE:  
4-94

**DTIC**